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#5 Priority
Paper
1-27-93
M. Co.

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Signed

M. Russell

Dated 7 September 1992

11 OCT 1991



15 OCT 1991 00280215

PAT 1 77 00

15.00

Your reference

2/H-36610/IMP.

0121657.2

Notes

Please type, or write in dark ink using CAPITAL letters. A prescribed fee is payable for a request for grant of a patent. For details, please contact the Patent Office (telephone 071-829 6910).

Rule 16 of the Patents Rules 1990 is the main rule governing the completion and filing of this form.

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The
Patent
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Request for grant of a Patent

Form 1/77

Patents Act 1977

1 Title of invention

- 1 Please give the title of the invention

LUBRICANTS

2 Applicant's details☐ **First or only applicant**

- 2a If you are applying as a corporate body please give:

Corporate name IMPERIAL CHEMICAL INDUSTRIES PLC

Country (and State) United Kingdom
of incorporation, if appropriate

- 2b If you are applying as an individual or one of a partnership please give in full:

Surname

Forenames

- 2c In all cases, please give the following details:

Address Imperial Chemical House, Millbank,
London

UK postcode SW1P 3JF
(if applicable)

Country United Kingdom

ADP number 935003
(if known)

2d, 2e and 2f: If there are further applicants please provide details on a separate sheet of paper.

☒ **Second applicant (if any)**

2d If you are applying as a corporate body please give:

Corporate name

Country (and State
of incorporation, if
appropriate)

2e If you are applying as an individual or one of a partnership please give in full:

Surname

Forenames

2f In all cases, please give the following details:

Address

UK postcode
(if applicable)

Country

ADP number
(if known)

③ An address for service in the United Kingdom must be supplied

Please mark correct box

③ Address for service details

3a Have you appointed an agent to deal with your application?

Yes ☒

No ☐ → go to 3b

↓
please give details below

Agent's name ROBERTS, JONATHAN WINSTANLEY

Agent's address Legal Department: Patents
Imperial Chemical Industries PLC
P O Box 6, Bessemer Road
Welwyn Garden City
Hertfordshire

Postcode AL7 1HD

Agent's ADP number 04432522001 ✓

3b: If you have appointed an agent, all correspondence concerning your application will be sent to the agent's United Kingdom address.

3b If you have not appointed an agent please give a name and address in the United Kingdom to which all correspondence will be sent:

Name

Address

Postcode

ADP number
(if known)

Daytime telephone
number (if available)

4 Reference number

- 4 Agent's or applicant's reference number (if applicable)

2/11. 36610/PA/P

5 Claiming an earlier application date

- 5 Are you claiming that this application be treated as having been filed on the date of filing of an earlier application?

Yes ☐

No ☒ → go to 6

↓
please give details below

- ☐ number of earlier application or patent number
- ☐ filing date

(day month year)

- ☐ and the Section of the Patents Act 1977 under which you are claiming:

15(4) (Divisional) ☐ 8(3) ☐ 12(6) ☐ 37(4) ☐

6 Declaration of priority

- 6 If you are declaring priority from previous application(s), please give:

Country of filing	Priority application number (if known)	Filing date (day, month, year)
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6 If you are declaring priority from a PCT Application please enter 'PCT' as the country and enter the country code (for example, GB) as part of the application number.

Please give the date in all number format, for example, 31/05/90 for 31 May 1990.

7 The answer must be 'No' if:

- any applicant is not an inventor
- there is an inventor who is not an applicant, or
- any applicant is a corporate body.

8 Please supply duplicates of claim(s), abstract, description and drawing(s).

7 Inventorship

7 Are you (the applicant or applicants) the sole inventor or the joint inventors?

Please mark correct box

Yes ☐

No ☒

A Statement of Inventorship on Patents Form 7/77 will need to be filed (see Rule 15).

8 Checklist

8a Please fill in the number of sheets for each of the following types of document contained in this application.

Continuation sheets for this Patents Form 1/77

Claim(s)

-

Description

5

Abstract

-

Drawing(s)

-

8b Which of the following documents also accompanies the application?

Priority documents (please state how many)

Translation(s) of Priority documents (please state how many)

Patents Form 7/77 - Statement of Inventorship and Right to Grant
(please state how many)

Patents Form 9/77 - Preliminary Examination/Search

Patents Form 10/77 - Request for Substantive Examination

9 You or your appointed agent (see Rule 90 of the Patents Rules 1990) must sign this request.

Please sign here →

9 Request

I/We request the grant of a patent on the basis of this application.

IMPERIAL CHEMICAL INDUSTRIES PLC

Signed

Authorised Officer

Date 11 11 1991

(day month year)

A completed fee sheet should preferably accompany the fee.

Please return the completed form, attachments and duplicates where requested, together with the prescribed fee to:

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The Patent Office
State House
66-71 High Holborn
London
WC1R 4TP**

Lubricants

This invention relates to lubricants and in particular their use in heat transfer devices.

Heat transfer devices of the mechanical recompression type such as those used in refrigerators, freezers, heat pumps and automobile air conditioning units are well known. In such devices a working fluid of a suitable boiling point evaporates at a low pressure taking heat from the surrounding zone. The resulting vapour is then compressed and passes to a condenser where it condenses and gives off heat to a second zone. The condensate is then returned through an expansion valve to the evaporator so completing the cycle. The mechanical energy required for compressing the vapour and pumping the fluid is provided by, for example, an electric motor or an internal combustion engine.

The working fluids used in these heat transfer devices include chlorine containing fluoroalkanes, e.g. dichlorodifluoromethane (R-12), chlorodifluoromethane (R-22) and mixtures thereof with for example fluoroalkanes, such as difluoroethane (R-152a). The use and production of such chlorine containing fluoroalkanes is likely to be severely limited by international agreement in order to protect the stratospheric ozone layer. It has been proposed that certain chlorine containing fluoroalkanes be replaced with for equivalent fluoroalkanes which have comparable boiling points and other thermal properties, but which also less damaging or benign to the stratospheric ozone layer, thus R-12 is generally being replaced by a new refrigerant, 1,1,1,2-tetrafluoroethane (R-134a). Unfortunately, the replacement refrigerants, notably R-134a, are insufficiently soluble in mineral oils to allow the latter to be used as lubricants. Consequently, numerous alternative lubricants such as polyalkylene glycols terminating in hydroxyl and other groups, esters of polyhydroxy alcohols with mono and polyfunctional acids, halo substituted esters and ethers have been proposed as lubricants for use with the replacement refrigerants.

Unfortunately, R-134a cannot be used to directly replace

other refrigerants such as R-22 and R-502 due to the different boiling characteristics and thermal properties of these other refrigerants. It has thus been proposed that these other refrigerants are replaced by refrigerant mixtures, in particular binary mixtures of refrigerants, such as R-134a and difluoromethane (R-32) or pentafluoroethane (R-125) and R-32. Unfortunately, these refrigerant mixtures are also not sufficiently soluble in mineral oils to allow the latter to be used as lubricants. Furthermore, knowledge of the miscibility and solubility of an alternative lubricant, which may be acceptable for use with R-134a, in one component of the mixture does not allow its solubility or miscibility in either an other component of the mixture or in the mixture itself to be determined, thus the effectiveness of an alternative lubricant for use with the refrigerant mixture has not hitherto been easily assessed.

It has now been found that if a prospective lubricant is at least partially soluble in each component of the refrigerant mixture then it will be at least partially soluble in the refrigerant mixture, thereby enabling its use as a lubricant with the refrigerant mixture, notwithstanding that it may be immiscible with one or more components or may be immiscible with the refrigerant mixture.

Accordingly the present invention provides a lubricant composition comprising

- (a) a working fluid comprising a mixture of at least two components, wherein each component is a hydrofluoroalkane or a fluoroalkane; and
- (b) sufficient to provide lubrication of a lubricant which is at least partially soluble in each component.

The working fluid may be a refrigerant mixture comprising two, three or more components. Suitably the components may be selected from known hydrofluoroalkanes and fluoroalkanes, for example R-134a, R-125, and R-32. Typically, a refrigerant mixture suitable to replace conventional R-22 may comprise (by weight) equal proportions of R-134a and R-32, or equal proportions

of R-125 and R32.

Suitable lubricants may be selected from those currently used with R-134a, provided that the requirement of at least partial solubility is met. Particularly effective lubricants are those selected from the classes known as polyalkylene glycols and neopentyl polyol esters, with neopentyl polyol esters being especially suitable due to their generally high level of thermal stability. Suitable neopentyl polyol esters include esters of tri, di and mono pentaerythritol, trimethylolpropane, trimethylolethane and neopentylglycol. Such esters may be formed with acids such as linear and/or branched aliphatic carboxylic acids, e.g. lower carbon length alkanolic acids, n-pentanoic through to n-decanoic acids, and branched alkanolic acids. Refrigeration systems which contain replacements for R-22 and R-502 typically operate at temperatures above those using R-134a as the sole refrigerant. Thus, it is desirable that the lubricant which is used in such a system is thermally stable at the elevated operating temperatures. Lubricants which are particularly stable at high temperatures include those comprising one or more esters formed from linear aliphatic carboxylic acids, or formed from a mixture of linear and branched aliphatic acids wherein a substantial proportion of the acids in the mixture are linear, e.g. at least 25 mol%, preferably at least 50 mol%, and particularly at least 75 mol%. Alternatively, a particularly thermally stable lubricant may comprise a mixture of esters comprising at least one linear ester formed from a linear aliphatic carboxylic acid and at least one branched ester formed from a branched aliphatic carboxylic acid, and wherein the linear ester is a substantial proportion of the mixture, e.g. at least 25 mol%, preferably at least 50 mol%, and particularly at least 75 mol%. Suitable polyalkylene glycols include hydroxyl group initiated polyalkylenes glycols, e.g. ethylene and/or propylene oxide polymers initiated on mono or polyfunctional alcohols such as methanol or pentaerythritol or glycerol. Such polyalkylene glycols may also be endcapped with suitable terminal groups, such

as alkyl, e.g. methyl groups.

The present invention is illustrated by reference to the following examples.

In these examples various compositions were prepared comprising 15% w/w of a lubricant and the complementary percentage of a refrigerant mixture comprising (by weight) equal proportions of R-134a and R-32. The lower miscibility temperature was determined, i.e. the lowest temperature at which the lubricant remained miscible with the refrigerant mixture. The results are displayed in Table 1, together with the lower miscibility temperature of the lubricant in each component of the refrigerant mixture. Each lubricant was at least partially soluble in the each of the constituents of the mixture and also in the mixture itself.

Table 1

Lubricant	Lower Miscibility Temperature (°C)		
	R-134a	R-32	Mixture
PE6	<-60	0	-27
"EMKARATE" RL 212	-25	>20	-3
"EMKARATE" RL 184	10	>20	>10
"EMKAROX" RL 118	<-60	I	I

"I" denotes immiscibility over the range -50°C to +20°C.

PE6 is an ester of pentaerythritol and n-hexanoic acid.

"EMKARATE" RL 212 and "EMKARATE" RL 184 are commercially available lubricants for use with R-134a comprising neopentyl polyol esters and are obtainable from ICI Chemicals and Polymers Ltd.

"EMKAROX" RL 118 is a commercially available lubricant for use with R-134a comprising an endcapped polyalkylene glycol and is obtainable from ICI Chemicals and Polymers Ltd.

("EMKARATE" and "EMKAROX" are trademarks of ICI Chemicals and Polymers Ltd).

In a further series of experiments various compositions were prepared comprising 15% w/w of each of the above lubricants and the complementary percentage of refrigerant R-125, in which each of the lubricants is at least partially soluble. The lower

miscibility temperature was again determined and the results are displayed in Table 2.

Table 2

Lower Miscibility Temperature

Lubricant

R-125

PE6

<-60

"EMKARATE" RL 212

<-60

"EMKARATE" RL 184

<-60


"EMKAROX" RL 118

<-60

CUSHMAN, DARBY & CUSHMAN

Inventor: CORR

Filed: 10/7/92

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